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A novel approach to fabricate porous alumina ceramics with excellent properties via pore-forming agent combined with sol impregnation technique

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ABSTRACT

An innovative approach for fabricating porous alumina ceramics (PACs) with improved mechanical and thermal properties using walnut shell powders as pore-forming agent combined with alumina sol impregnation is reported in the present work. It is demonstrated that uniform distribution of spherical pores can be observed in as-prepared PACs by using above technical route. The decrease of walnut shell powder sizes significantly promotes the enhancement of crushing strength and reduction of thermal conductivity of the PACs. Meanwhile, the impregnated alumina sol is favoring for the formation of spherical micro-pores, then further improves their mechanical and thermal insulation performances. The lowest thermal conductivity and highest crushing strength of resulting sample reach 0.16 W/m K and 29.2 MPa, respectively. This novel method offers new possibilities to fabricate high-quality PACs.

1. Introduction

Porous alumina ceramics (PACs) are used widely in filtration, thermal insulators and catalyst fields due to their low thermal conductivity, high melting point and chemical stabilities [1–5]. As for furnace thermal insulators, alumina hollowsphere bricks are currently applied owing to their excellent mechanical properties. However, the relative high thermal conductivity and energy-intensive fabrication process make them unable to satisfy the ever-increasing request of modern industry [6,7]. Therefore, it is of great importance to develop a novel and low-cost PACs with excellent insulation performance and mechanical strength simultaneously.

Thus far, various methods are employed for preparing alumina or alumina-based ceramics, including foam-gelcasting [8], sacrificial templating [9,10] and pore-forming agent (PFA) [11–13]. The last method is widely used to fabricate porous samples with high porosity or complex shape because of its simple preparation procedure and high controllability [14,15]. In addition, the pore structures of porous ceramics are intrinsically relate to the morphology and elasticity of PFA, which would largely affect their performances. The traditional PFA employed to prepare PACs mainly including sawdust [16], rice husk [17] and polymethylmethacrylate (PMMA) microspheres [18], etc. Nevertheless, these traditional PFA present some obstacles in preparing porous products with less defect and good materials homogeneity due to the considerable spring back effect caused by their high elasticity [19,20]. Furthermore, the impurities after burn-out may also deteriorate the high-temperature performances of porous ceramics. Other than traditional high elasticity PFA, rigid PFA including ceramic hollow microspheres [21,22] and other particles [23], etc., have been developed to prepare high performance porous ceramics. In comparison with high elasticity PFA, rigid PFA exhibits many advantages, namely low elasticity, low toxicity and effective compatibility with ceramic paste. Nevertheless, low porosity (about 45–60%) and complex preparation processing are the drawbacks to limit their further application, thus many researchers have focused on the exploitation of facile, low cost, and eco-friendly fabrication method in recent years [24,25].

Walnut shell powder is a natural organic matter possessing non-toxicity after burning and significant level of organics, which has been widely applied in the filterable, medical and catalytic fields [26,27]. However, the irregular pores after burn-out still present a challenge for its wider application. To address this problem, in the present work, four different sizes of walnut shell powders were introduced as the PFA of PACs. Based on our previous work [28], alumina sol was introduced in this work during the preparation of PACs to modify the pore structure

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Table 1
Element compositions of walnut shell powders.

Walnut shells	C	O	Si	K	Na	Ca	Other
Mass fraction	64.38	33.85	0.164	0.532	0.140	0.197	0.737

and improve the performance of PACs. The effects of walnut shell powders sizes on the pore size, pore proportions and pore morphology were investigated systematically. Also, the roles of pore structures on thermal conductivity and mechanical properties of PACs were analyzed.

2. Experimental procedures

2.1. Preparation of PACs

Tabular alumina (≤ 74 μm and ≤ 45 μm, Qingdao, China), α-Al_2O_3 (2 μm, Qingdao, China) and ρ-Al_2O_3 (5 μm, Zhengzhou, China) were used as starting materials in this work. Four different sizes of walnut shell powders (WS, 45 μm; 22 μm; 13 μm and 7 μm, Shijiazhuang, China) were chosen as PAF and their added volume percentage were almost same to ensure the same porosity of fired samples. The chemical compositions of walnut shell powder were listed on Table 1. Poly vinyl alcohol (PVA), one in liquid form (5 wt% solid content, Beijing, China) and commercial alumina sol (5 wt% solid content, Handan, China) were adopted as binder and coating compositions. Fig. 1 gives the microstructures of walnut shell powders and alumina sol particles. As can be seen that the walnut shell powders exhibit irregular morphology with various sizes, and the alumina sol present perfect spherical particles. Fig. 2 illustrates the detailed process for fabricating the PACs. Firstly, the ceramic pastes consisting of tabular alumina, α-Al_2O_3, walnut shell powders and deionized water were added into 5 wt% PVA aqueous solution. The obtained pastes were then put into cold isostatic pressing mold and pressed under 5 MPa. The green bodies are pre-fired at 1400 °C at a rate of 5 °C/min for 2 h. Most of the pre-fired samples were immediately heat up to 1600 °C at a rate of 10 °C/min. The other pre-fired samples were sintered beforehand impregnating alumina sols under vacuum followed by drying, as described above. Altogether, five different types samples were obtained by varying the wall shell powder sizes and whether the as-fired compositions were impregnated beforehand or not (i.e., WS45: 45 μm; WS22: 22 μm; WS13: 13 μm; WS7: 7 μm and WS7-5: 7 μm, 5 wt%).

2.2. Characterization methods

Apparent porosity of the obtained alumina samples was measured according to Archimedes method by distilling deionized water. Total porosity was calculated according to Eq. (1):

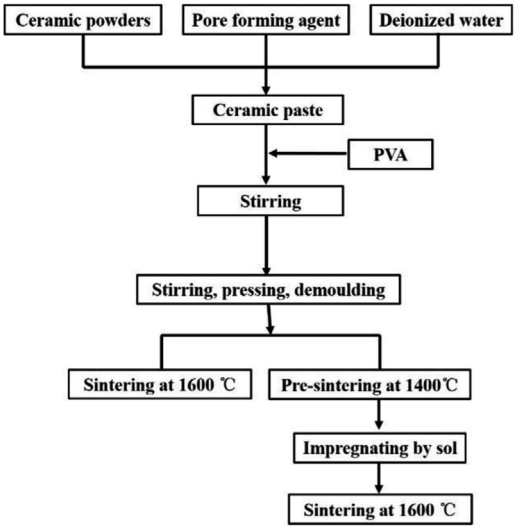

Fig. 2. Scheme depicting the experimental procedure for the porous alumina ceramics production.

$$V_{total} = 1 - \frac{\rho_{bulk}}{\rho_{true}} \quad (1)$$

here, V_{total} was the total porosity, ρ_{bulk} and ρ_{true} were the bulk density and true density of porous alumina ceramics, respectively. Close porosity of the obtained alumina samples was calculated according to Eq. (2):

$$V_{close} = V_{total} - V_{open} \quad (2)$$

where V_{close} was the close porosity, V_{total} and V_{open} were total porosity and open porosity of sintered samples, respectively. Microstructures were characterized with a scanning electron microscopy (SEM, Carl Zeiss, Jena, Germany). Pore size distribution was characterized by Mercury Intrusion Porosimetry (Autopore IV9500, Micromeritics Instrument Corp., USA). Pore shape factor was calculated according to Ref. [29]. Thermal conductivity with dimensions of Φ180 mm × 20 mm were characterized using a thermal constants apparatus (PBDR-02, Precondar, PR China). Cold crushing strength was determined using cylindrical samples of Φ30 mm × 20 mm according to the GB-T 3997.2-1998. More than fifteen samples were measured to obtain the average value. Based on the crushing strength result, Weibull distribution plots were evaluated as follows:

$$\ln\ln[1/(1-F)] = m\ln\sigma_n - m\ln\sigma_0 \quad (3)$$

where m was Weibull modulus, σ_n and σ_0 were the crushing strength of the nth bar and characteristic crushing strength, respectively.

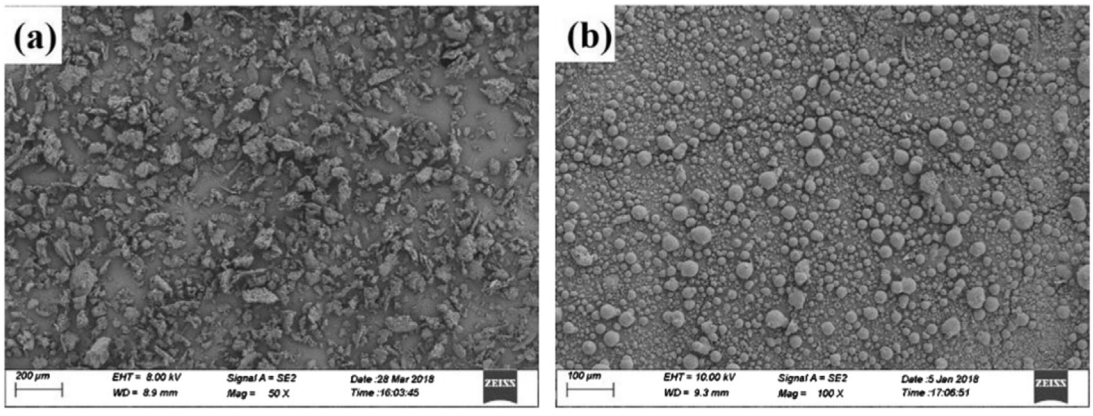

Fig. 1. SEM micrographs of (a) walnut shell powders and (b) alumina sol particles.